IN THE UNITED STATES

PATENT AND TRADEMARK OFFICE

APPLICANTS:	Leonid Grigorian, Loius Hornyak, Anne Dillon, and Michael J. Heben.
SERIAL NO.:	09/825,870
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TITLE:	CHEMICAL VAPOR DEPOSITION GROWTH OF SINGLE-WALL CARBON NANOTUBES
EXAMINER:	Peter J. Lish
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CERTIFICATE OF MAILING

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Dated: Nov. 1/2005 By: Narinder Banait

Narinder S. Banait, Reg. No.: 43,482

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APPEAL BRIEF (37 C.F.R. 1.192)

Real Parties in Interest

The subject application is owned by HONDA MOTOR CO., LTD., of Tokyo, Japan, and Midwest Research Institute, of Kansas City, Mo.

Related Appeals and Interferences

There are no known related appeals or interferences that may directly affect, be directly affected by, or have a bearing on the Board's decision in the pending appeal.

Status of Claims

Claims 1-18 stand finally rejected. On June 1, 2005, the appellants appealed from the final rejection of claims 1-18. The claims on appeal are set forth in an Appendix I attached hereto.

Status of Amendments

The appellants have not amended the claims since the final rejection.

Summary of the Invention

Appellants' invention provides methods for the preparation of single-wall carbon nanotubes (SWNTs). (Spec. paragraph 12.) A single-wall carbon nanotube is defined as a molecule formed primarily from sp^2-hybridized carbon atoms bound together in the shape of a hollow tube that is capped at each end. The typical SWNT is made of graphite sheet capped with half a fullerene molecule on each end, and with a diameter of less than about 3 nm (Spec. paragraph 42).

The applicants' invention relates to chemical vapor deposition for the preparation of SWNTs from methane with a growth temperature of about 670 °C to about 800 °C and gas pressure of about 400 torr to about 600 torr. Appellants' claimed invention comprises

"decomposing the methane gas composition in the presence of the supported iron-containing catalyst, under a gas pressure of about 400 torr to about 600 torr and for a time sufficient to grow single-wall carbon nanotubes at a temperature from about 670° C to about 800° C" (Claim 1), "decomposing the methane gas composition in the presence of the Al_2O_3/Fe/Mo catalyst, under a gas pressure of about 400 torr to about 600 torr and for a time sufficient, to grow single-wall carbon nanotubes at a temperature from about 670° C to about 800° C" (Claim 9), or "decomposing the methane gas composition in the presence of the Al_2O_3/Fe/Co/Mo catalyst, under a gas pressure of about 400 torr to about 600 torr and for a time sufficient, to grow single-wall carbon nanotubes at a temperature from about 680° C to about 800° C" (Claim 13). As appellants' specification teaches, higher yields of SWNTs are achieved within the growth temperature, and the production of amorphous carbon and other side products are decreased. Further, the yield of SWNTs outside of the growth temperature range is said to drop dramatically (Spec. paragraphs 12, 45, 50, 51, and 75).

Issues

The issues for appeal are whether claims 1-4 and 6-8 are properly rejected under 35 U.S.C. section 103(a) as being unpatentable over U.S. Patent No. 5,973,444 to Xu *et al.*, filed November 12, 1998 ("Xu"), and Claims 5 and 9-18 are properly rejected under 35 U.S.C. section 103(a) as being unpatentable over Xu in view of U.S. Patent No. 6,413,487 to Resasco *et al.* filed June 2, 2000 ("Resasco").

Grouping of Claims

For the purposes of the specific arguments made by the appellants in the present appeal, claims 1-18 stand or fall separately.

Argument

I. <u>Xu fails to teach or suggest every element of the claims</u>

To establish prima facie obviousness of a claimed invention, all the claim limitations must be taught or suggested by the prior art. *In re* Royka, 490 F.2d 921, 180 USPQ 580 (CCPA 1974). MPEP § 2143.03. The § 103 rejection of claims 1-4 and 6-8 over Xu is improper because Xu discloses the preparation of carbon fibers only, not SWNTs. Further, the examiner has failed to identify any motivation to use the method for preparing carbon fibers disclosed by Xu to prepare SWNTs.

The applicants define single-wall carbon nanotubes (SWNTs) (Spec. paragraph 12, page 8, line 31 to page 9, line 4) as a molecule formed primarily from sp^2-hybridized carbon atoms bound together in the shape of a hollow tube that is capped at each end. Further, SWNTs are said to be one atomic carbon layer in thickness with a diameter of less than about 3 nm. Xu describes the structure of the carbon fiber emitters at column 9, lines 25-61. Xu states that straight fibers are grown at high temperatures, whereas vermicular fibers, having irregular twisting structures and amorphous structure, are usually produced at temperatures below about 900 °C. Further, the carbon fiber emitters are said to have an average diameter less than about 2-10 microns, most preferably in the range of about 20 nm to about 200 nm.

Thus, SWNTs are hollow whereas carbon fibers are solid; SWNTs have defined structure whereas carbon fibers can have amorphous structure, graphite like structure, and herringbone structure; and the diameter of SWNTs (less than 3 nm) is significantly smaller than the diameter of carbon fibers (20 nm to 200 nm). Thus, the SWNTs produced by the applicants and the carbon fibers disclosed by Xu *et al.* are not the same. Carbon fibers and SWNTs have different structures and different properties, and the art recognizes SWNTs and carbon fibers to be different and not as equivalents.

The examiner states: "However, Xu et al. specifically states that the process may produce, amongst other things, 'single-wall tubular structures'." Advisory Action mailed 04/19/2005 at p. 2. The examiner incorrectly equated "single-wall tubular structures" with SWNTs. Xu describes the various morphologies of the fibers at column 9, lines 40-52, and states: "The fibers include single wall or multiple-walled tubular structures." The single-wall tubular structures of Xu are fibers. The applicants' were unable to find any disclosure of SWNTs in Xu.

The § 103 rejection of claims 1-4 and 6-8 is improper because Xu fails to teach or suggest the "single-wall carbon nanotubes" element of claims.

II. The ranges claimed are critical

The examiner rejected claims 1-4 and 6-8 under 35 U.S.C. § 103(a) as being unpatentable over Xu because the pressure and temperature ranges claimed by the applicants are encompassed within the pressure and temperature ranges disclosed by Xu. Advisory Action mailed 04/19/2005 at p. 2, and Final Office Action, mailed 12/01/2004 at p. 3, lines 7-11. The applicants claim a gas pressure of about 400 torr to about 600 torr and a temperature from about 670° C to about

800° C to grow SWNTs. Xu discloses a gas pressure of one millitorr to several atmospheres (column 8, lines 61-63), and a temperature range from about 300° C to about 1200° C (column 9, lines 3-13) to grow carbon nanowires. The examiner cites In re Malagari 182 USPQ 549 (CCPA, 1974) to support the obviousness rejection based on overlapping ranges. The applicants disagree because their disclosure provides evidence supporting the advantages of the claimed ranges, and provides evidence showing SWNTs cannot be prepared outside the claimed ranges, such as the pressure and temperature ranges of Xu.

> "The law is replete with cases in which the difference between the claimed invention and the prior art is some range or other variable within the claims. . . . In such a situation, the applicant must show that the particular range is critical, generally by showing that the claimed range achieves unexpected results relative to the prior art range." *In re Woodruff*, 919 F.2d 1575, 16 USPQ2d 1934 (Fed. Cir. 1990).

The applicants characterize SWNTs using Raman spectroscopy (Spec. paragraphs 43-45), specifically Raman signals at about 1593 and 1567 cm^{-1}. The signal at these wavelengths is said to increase in intensity as the purity and the percent of SWNT material increases. The data provided by the applicants in the specification demonstrated that SWNT growth occurred only within a window, as claimed.

Applicants' examples 2 and 4-7 and Figure 5 shows that no detectable Raman bands at temperatures less than 670 °C and Raman bands almost disappear at about 1000 °C. The data thus demonstrate SWNTs are not produced at temperatures less than 670 °C or above 1000 °C. The conclusion is further supported by TEM images shown in Figures 1 and 2 taken from SWNTs deposited at 700 °C to about 1000 °C. Further, the results illustrated in Figures 6 and 7 show that carbon deposit produced at temperatures greater than 800 °C consists mainly of amorphous carbon and not SWNTs. Thus, even though the pressure and temperature ranges

claimed by the applicants fall within the ranges disclosed by Xu, the data in the applicants' disclosure demonstrates the impossibility of producing SWNTs over Xu's ranges. At the time of the invention, the applicants were not aware of any publication that disclosed SWNTs could be produced across such a large range of pressures and temperatures. The experimental data provided by the applicants in their specification demonstrates that the ranges disclosed by Xu will certainly not produce SWNTs.

Accordingly, claims 1-4 and 6-8 are patentable over the cited art.

III. The combination of Xu with Resasco fails because Resasco teaches away from the invention

The examiner rejected Claims 5 and 9-18 under 35 U.S.C. section 103(a) as being unpatentable over Xu in view of Resasco. Xu is said not to teach the use of iron-molybdenum catalysts, but Resasco is said to teach bimetallic catalysts containing Group VIII metal and Group VIb metal. The applicants disagree because Resasco specifically excludes the use of Fe in the catalysts, therefore, one of skill in the art would not combine Xu with Resasco.

The applicants' claim 5 pertains to iron-containing catalyst Al_2O_3/Fe/Mo, independent claim 9 pertains to Al_2O_3/Fe/Mo catalyst, and independent claim 13 pertains to Al_2O_3/Fe/Co/Mo catalyst. Thus, the catalysts of claims 5 and 9-18 all contain iron. Iron is a Group VIII metal. However, Resasco explicitly excludes the use of iron in the bimetallic catalysts. Resasco, at column 7, lines 50-52, states the bimetallic catalyst "in a preferred version excluded Fe." Further, Resasco at column 7, lines 14-27 lists Co, Ni, Ru, Rh, Pd, Ir, and Pt as the Group VIII metals from which one of the bimetallic could be selected. The list excludes Fe, therefore, Fe is

not one of the Group VIII metals from which one of the bimetallic could be selected. Resasco then provides 21 specific examples of bimetallic catalysts which could be employed in their invention. Again, a bimetallic catalyst containing Fe is missing. Thus, Resasco clearly excluded Fe as an acceptable Group VIII metal for use in the bimetallic catalysts, and a skilled artisan would not substitute Fe for a Resasco Group VIII metal. In contrast, the catalysts of claims 5 and 9-18 all contain iron.

Resasco teaches away from using an iron-containing bimetallic catalysts, therefore, one of skill in the art would not combine Xu with Resasco. The § 103 rejection of claims 5 and 9-18 over Xu combined with Resasco is therefore improper.

Summary

For the foregoing reasons, appellants believe that the examiner's rejection of claims 1-18 were erroneous, and reversal of his decision is respectfully requested.

Respectfully submitted,
GRIGORIAN, ET AL.

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By: Narinder Banait

Narinder S. Banait, Reg. No.: 43,482
Attorney for Applicants
Fenwick & West LLP
801 California Street
Mountain View, CA 94041
Telephone: 650-335-7818
Facsimile: 650-938-5200
E-mail: nbanait@fenwick.com

Appendix I: Claims Involved in Appeal

1. A chemical vapor deposition process for the preparation of a single-wall carbon nanotube, comprising:

providing a methane gas composition and a supported iron-containing catalyst to a chemical vapor deposition chamber, and

decomposing the methane gas composition in the presence of the supported iron-containing catalyst, under a gas pressure of about 400 torr to about 600 torr and for a time sufficient to grow single-wall carbon nanotubes at a temperature from about 670° C to about 800° C.

2. A process of claim 1, wherein said temperature is from about 670°C to about 750°C.

3. A process of claim 1, wherein said temperature is from about 670°C to about 700°C

4. A process of claim 1, wherein said supported iron-containing catalyst is selected from the group consisting of: Al_2O_3/Fe/Mo/Co, Al_2O_3/ Fe/Mo, Al_2O_3/Fe/Co, Al_2O_3/Fe, and mixtures thereof.

5. A process of claim 4, wherein the supported iron-containing catalyst is Al_2O_3/Fe/Mo catalyst, and wherein the catalyst has a molar ratio of Al_2O_3:Fe:Mo of about (10-20) : 1 : $^{1}/_{3}$.

6. A process of claim 1, wherein said methane gas composition is methane or a mixture of methane and a carrier gas.

7. A process of claim 6, wherein said carrier gas is selected from the group consisting of: argon, nitrogen, helium, and mixtures thereof.

8. A process of claim 7, wherein said methane gas and said carrier gas are used in a ratio of about 1:1 by volume to about 1:10 by volume.

9. A chemical vapor deposition process for the preparation of single-wall carbon nanotubes, comprising:

providing a methane gas composition and an Al_2O_3/Fe/Mo catalyst to a chemical vapor deposition chamber, and

decomposing the methane gas composition in the presence of the Al_2O_3/Fe/Mo catalyst, under a gas pressure of about 400 torr to about 600 torr and for a time sufficient, to grow single-wall carbon nanotubes at a temperature from about 670° C to about 800° C,

wherein said single-wall carbon nanotubes have a diameter distribution ranging from about 0.7 nm to about 2.1 nm.

10. A process of claim 9, wherein the Al_2O_3/Fe/Mo catalyst has a molar ratio of Al_2O_3:Fe:Mo of about (10-20) : 1 : $^{1}/_{3}$.

11. A process of claim 9, wherein said temperature is from about 670 °C to about 750°C.

12. A process of claim 9, wherein said temperature is from about 670°C to about 700°C.

13. A chemical vapor deposition process for the preparation of single-wall carbon nanotubes, comprising:

providing a methane gas composition and an Al_2O_3/Fe/Co/Mo catalyst to a chemical vapor deposition chamber, and

decomposing the methane gas composition in the presence of the Al_2O_3/Fe/Co/Mo

catalyst, under a gas pressure of about 400 torr to about 600 torr and for a time sufficient, to grow single-wall carbon nanotubes at a temperature from about 680° C to about 800° C

wherein said single-wall carbon nanotubes have a diameter distribution ranging from about 0.7 nm to about 2.1 nm.

14. A process of claim 13, wherein the Al_2O_3/Fe/Co/Mo catalyst has a molar ratio of Al_2O_3:Fe:Co:Mo of about (10-20) : 1 : 0.23 : $^1/_6$.

15. A process of claim 13, wherein the Al_2O_3/Fe/Co/Mo catalyst has a molar ratio of Al_2O_3:Fe:Co:Mo of about (10-20) : 1 : 0.23 : $^1/_{18}$.

16. A process of claim 13, wherein the Al_2O_3/Fe/Co/Mo catalyst has a molar ratio of Al_2O_3:Fe:Co:Mo of about (10-20) : 1 : 0.23 : $^1/_{36}$.

17. A process of claim 13, wherein said temperature is from about 680 °C to about 750°C.

18. A process of claim 13, wherein said temperature is from about 680°C to about 700°C.